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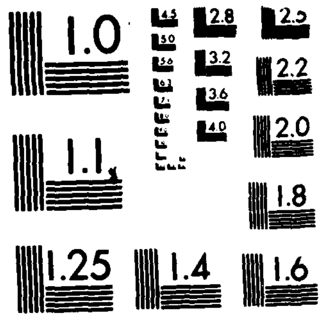
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THE OPTICAL SPECTRA OF AEROSOLS

Principal investigator: F.Borghese

Contractor: F.Borghese

Contract n° DAJA37-81-C-0895

Sixth Periodic Report

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On the basis of previous theory aimed at calculating the macroscopic optical constants of monodisperse model aerosol, it is now possible to calculate the absorption coefficient of a polydisperse aerosol, as well as the changes induced on it by changes in the structure of the scattering particles. This report consists of a summary of the mathematical development.

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The work we started about one year ago to study the collective properties of reliable model aerosols has given in the last semester, very interesting positive results. In fact on the basis of our previous theory aimed at calculating the macroscopic optical constants of a monodisperse model aerosol, we were able to calculate the absorption coefficient of a polydisperse aerosol and the changes induced on it by changes in structure of the scattering particles. The preliminary results have been accepted as an original contribution to the First Int. Aerosol Conf. to be held in Minneapolis (Minnesota) next september. The extended abstract, to be published in the proceeding of the Conference, is enclosed here to illustrate the methods and purposes of the work. A full lenght paper on the subject is now on the way of completition. I am sorry we are no able to enclose at present some figures to improve the understanding of our results. However, during the writing of the paper some questions arose that can be answered only through further calculations and a thorough examination of the previous obtained computer output. In this connection it is usefull to point out that the core storage required to get well converged results exceeds 4Mbytes; alternatively the storsge requirements can be considerably decreased by transforming some of the subroutines of our main program into functions. The cost to be paied for this transformation is an increase of the CPU time by a factor of 5 or 6. Anyway this seems to be the only practible way to treat aerosol composed of large particles. For this reason this transformation is at present under way.

To end this report I am glad to infor you that our

paper:

F.Borghese, P.Denti, R.Saija, G.Toscano and O.I.Sindoni

"Macroscopic optical constants of a cloud of randomly oriented nonspherical scatterers"

has been accepted for publication in Il Nuovo Cimento and since the proofs have already been corrected it is scheduled to be printed in next July.

Effect of the "chemical reactions" on the macroscopic optical constants of a model aerosol(*)

by

R.Saija(+), O.I.Sindoni(°), G.Toscano(+), F.Borghese(+§), P.Denti(+§) .

(*) Based on work supported by the U.S. Army European Research Office through Contract DAJA37-81-C-0895 and Grant DAJA45--84-C-0005.

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It is well known that the refractive index of a low-density dispersion of scatterers is a matrix whose (complex) elements are given by (R.G.Newton,"Scattering theory of waves and particles", McGraw-Hill, New York, 1966)

$$\mathcal{N}_{\eta'\eta} = \delta_{\eta'\eta} + \frac{2\pi}{Vk^2} \sum_{\nu} f_{\nu,\eta'\eta} \qquad (1)$$

In eq.(1) k is the magnitude of the wavevector of the incident light and ν numbers the scatterers within the volume V. Furthermore, if $\underline{u}_{\eta}$, is the polarization vector of the incident light, $f_{\nu,\eta'\eta}$ is related to the normalized forward-scattering amplitude, for polarizzation η, of the ν-th scatterer, through

$$f_{\nu,\eta'\eta} = \underset{\sim}{u}^{*}_{\eta'} \cdot \underset{\sim}{f}_{\nu,\eta} . \qquad (2)$$

The elements $\mathcal{N}_{\eta'\eta}$ are related to the macroscopic refractive index, n_{η}, and to the absorption coefficient, γ_{η}, for polarization η, through

$$n_{\eta} = \mathrm{Re}(\mathcal{N}_{\eta\eta}) \quad , \quad \gamma_{\eta} = 2k\,\mathrm{Im}(\mathcal{N}_{\eta\eta})$$

respectively. We recall that if all the scatterers are identical and spherically symmetric, $f_{\nu,\eta'\eta}$ does not depend on ν, while if the scatterers are identical but nonspherical, $f_{\nu,\eta'\eta}$ depends on their orientation with respect to the incident field.

In a preceding paper (F.Borghese, P.Denti, R.Saija, G.Toscano and O.I.Sindoni, 'Macroscopic optical constants of a cloud of randomply oriented scatterers', Nuovo Cimento, to be published,1984) we proposed to account for the lack of spherical symmetry of some kind of scatterers by modeling them as clusters of spheres, and, making full use of the features of the model, succeeded in factorizing $f_{\nu,\eta}$ into a part depending only on the structure and a part depending only on the orientation of the clusters. For a dispersion of identical clusters we were then able to sum even the orientations and to express $\mathcal{N}_{\eta'\eta}$ in terms of the multipolar amplitudes scattered by a cluster whatever. The results is

$$\mathcal{N}_{\eta'\eta} = \delta_{\eta'\eta} + \frac{N}{4k^2 i} \sum_{LM} \frac{1}{2L+1} W^*_{\eta' LM} W_{\eta LM} \times$$

$$\times \sum_{M'} \left(\bar{U}^{(A)}_{\eta LMLM'} + \eta' \, \bar{U}^{(B)}_{\eta LMLM'} \right) \qquad (3)$$

where the $\bar{U}$'s depend only on the structure of the clusters, whose number density is N, and $W_{\eta LM}$ is proportional to the multipolar amplitudes of the incident plane wave field. Eq.(3) can be immediately extended to the dispersions of more than one kind of clusters. If $N_\alpha = N c_\alpha$ is the number density of the α-th kind of clusters, we can write

$$\mathcal{N}_{\eta'\eta} = \delta_{\eta'\eta} + N \sum_{\alpha, L} c_\alpha \, \xi_{\eta'\eta L} \, S_{\alpha\eta'\eta L} \tag{4}$$

where we define

$$\xi_{\eta'\eta L} = \frac{1}{4\pi i} \, \frac{1}{2L+1} \sum_M W^*_{\eta' L M} \, W_{\eta L M}$$

$$S_{\alpha\eta'\eta L} = \frac{1}{k^3} \sum_{M'} \left(\bar{U}^{(1)}_{\alpha\eta L M L M'} + \eta' \, \bar{U}^{(2)}_{\alpha\eta L M L M'} \right)$$

The index α added to the $\bar{U}$'s individuate the quantities appropria te to the α-th kind of cluster.

The cluster model and eq.(4) lend themselves to the calculation of the changes induced on the optical constants of the dispersion by structural rearrangements of the clusters or by the redistribution of the spheres among different clusters; these rearrangements should account for the occurrence of "chemical reactions" among the scattrers of the dispersion. In fact, eq.(4) can be used for different values of the relative concentrations, c_α, of the reagents and of the reaction products, subject to the costraint:

$$\sum_\alpha c_\alpha = 1$$

of course. The results of the calculations are well described

the quantity:

$$\Gamma = [p\gamma^{(f)} + (1-p)\gamma^{(i)}]/\gamma^{(i)}$$

where $\gamma^{(f)}$ and $\gamma^{(i)}$ are the absorption coefficients of the dispersion of the reaction products and of the reagents, respectively, and p is the completion parameter of the reaction. The parameter Γ has been computed for a few "reactions" chosen so as to evidence the effect of the rearrangement on the spheres. In all cases considered the changes in the absorption coefficient are more visible when dramatic changes of the structure of the clusters occur. Of course, the results we present here are to be considered as preliminary ones and do not pretend, at this stage, to fit any actual experimental data. In any case our work shows that the anysotropy of the scatterers plays a relevant role in the process of the coherent electromagnetic propagation and that, neglecting the possible structural changes of the scatterers may prevent any realistic interpretation of the experimental data.

Annex

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